

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of : Hongyuan WANG et al.

Serial Number : 10/584,189

Filed : June 23, 2006

For : Aromatic polyamic acid and polyimide

Group Art Unit : 1796

DECLARATION UNDER 37 CFR 1.132

Honorable Commissioner for patents

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Sir:

Now comes Hongyuan WANG who declares and says that:

1. I am a co-inventor of the United States Patent Application Serial Number 10/584,189.
2. I graduated from Shanghai Jiaotong University (China), Faculty of Engineering, department of Industrial Chemistry in 1992, and I was awarded the doctor's degree from Yamaguchi University (Japan) in 1998.
3. I have been employed by Nippon Steel Chemical Co., Ltd. since 2001, and I investigated the syntheses and development of novel aromatic functional polyimide and alkoxyated benzidine monomers for the electrical and optical fields of applications.
4. I have synthesized the polyimide described by Harris in U. S. Patent No.

5,071,997, determined its properties, and compared them with those of the polyimide described in the specification of this application (Patent Application Serial Number 10/584,189).

I hereby prove that the polyimide according to this invention produces effects unpredictable by the invention of Harris. Further, I prove that the polyimide relating to this invention cannot easily be arrived at from the account given in the invention of Harris.

EXPERIMENT

Unless otherwise noted, raw materials and the measurement condition in the experiment are used in the same way as raw materials and the measurement condition used in the example described in the specification of this application and the measurement condition.

The following abbreviation was used for certain components used in the experiments:

PMDA: pyromellitic dianhydride

BTMB: 2,2'-bis(trifluoromethyl)benzidine

m-NPOB: 2,2'-di-n-propyloxybenzidine

m-PHOB: 2,2'-diphenyloxybenzidine

DMAc: N,N-dimethylacetamide

Experiment 1-3

To synthesize each of polyamic acids E, J and R, a diamine shown in Table A was dissolved into 180g of DMAc as a solvent while being stirred in a 300-ml separable flask in a stream of nitrogen. Next, a tetracarboxylic dianhydride shown in Table A was added. After that, the solution was subjected to a polymerization reaction while being continuously stirred at room temperature

for 3 hours, whereby a yellow-to-tan viscous solution of each of the polyamic acids E, J and R each serving as a polyimide precursor was obtained. The viscosity of the polyamic acids thus obtained is shown in Table A. The viscosity was determined with a cone and plate viscometer equipped with a constant-temperature bath (E type viscometer manufactured by Tokimec Inc.) at 25°C.

Each of the polyimide precursor solutions E, J and R was applied to an electrodeposited copper foil with a surface roughness (R_z) of 0.3 μm with an applicator in such a manner that the film thickness after drying would be about 15 μm , and was then dried at 130 °C for 2.4 minutes. After that, the resultant was subjected to a stepwise heat treatment at each of 130°C, 160°C, 200°C, 230°C, 280°C, 320°C, and 360°C for 2 to 30 minutes, whereby a polyimide layer was formed on the copper foil.

The laminate obtained above was processed to form a circuit by etching the copper foil off with an aqueous solution of ferric chloride while leaving a 1 mm-wide line of the copper foil. Thereafter, the resin side of the 1 mm-wide laminate was fixed to an aluminum plate with a double-faced adhesive tape and, with the aid of a tension tester, the copper foil was peeled in a 180° direction at a rate of 50 mm/min to determine the peel strength.

Furthermore, the copper foil was removed from the laminate through etching by using an aqueous solution of ferric chloride, whereby each of film-like polyimides E, J and R was produced. Then, the glass transition temperature (T_g), coefficient of moisture absorption, and coefficient of humidity expansion (CHE) of each polyimide were determined. The polyimides E, J and R mean that they were obtained from the polyamic acids E, J and R. Table B shows the results.

【Table A】

	Experiment 1	Experiment 2	Experiment 3
BTMB (g)			11.8727
PHOB (g)		12.6060	
NPOB (g)	2.0363		
PMDA (g)	1.4637	7.3940	8.1273
DMAc (g)	31.50	180	180
solid content (wt%)	10	10	10
molar ratio	0.990	0.990	1.005
viscosity (cP)	3353	10444	2611

【Table B】

	Experiment 1	Experiment 2	Experiment 3
polyamic acid	E	J	R
polyimide film	E	J	R
peel strength (kN/m)	0.36	0.37	0.08
Tg (°C)	378	394	388
coefficient of moisture absorption (wt%)	0.64	0.55	0.55
CHE (ppm/%RH)	2.4	3.6	5.3

Remarks

1) In Experiments 1-3, the polyamic acids and polyimides were synthesized using common raw materials except for the diamines under common reaction conditions for the purpose of making a precise comparison. The solid content was matched to that of Example 1 described by Harris (U. S. Patent No. 5,071,997).

2) In Experiments 1-2, the polyamic acids and polyimides corresponding to those in Example 5 and 10 of this application were synthesized, their properties were determined, and the results are shown. Polyamic acid E and polyimide E in Experiment 1 and polyamic acid J and polyimide J in Experiment 2 respectively exhibit the same properties (in respect to Tg, coefficient of moisture absorption, and CHE) as those of polyamic acid E and polyimide E in Example 5 and of polyamic acid J and polyimide J in Example 10 of this application.

3) In Experiment 3, polyamic acid R and polyimide R corresponding to those in Example 1 described in the invention of Harris (U. S. Patent No. 5,071,997) were synthesized, their properties were determined, and the results are shown.

Conclusions

1) A comparison of Experiments 1-2 and Experiment 3 indicates that, in spite of the fact that each polyimide exhibits roughly the same Tg, coefficient of moisture absorption, and CHE, the peel strength of the polyimides in Experiments 1-2 is roughly five times that of the polyimide in Experiment 3.

2) Harris describes that his polyimides show good solubility, but this is another way of saying that his polyimides show poor solvent resistance. On the other hand, a film of polyimide E in Experiment 1 and a film of polyimide J in Experiment 2 show no change when immersed in a strongly alkaline etching

solution at 80°C for 1 hour. The strongly alkaline etching solution here is composed of 33.5 wt% of H₂O, 33.5 wt% of KOH, 22.0 wt% of ethylene glycol, and 11.0 wt% of ethylenediamine.

3) From the results described above, it can be said that the polyimides of this invention are comparable in resistance to heat, thermal expansion, and moisture absorption to, but far superior in adhesive properties and chemical resistance (solvent resistance) to the polyimides disclosed by Harris.

4) In conclusion, the polyamic acids and polyimides of this invention can be said to produce effects which are not expected from those disclosed by Harris.

5) According to Harris (U. S. Patent No. 5,071,997), the group A in formula (I) denotes halogen, alkyl, substituted alkyl, nitro, cyano, thioalkyl, alkoxy, aryl, or substituted aryl and they are comparable in their effect. However, as demonstrated by Experiments 1-3, the alkoxy and substituted alkyl groups bring about large changes in the properties of polyimides.

Harris shows only one kind of substituted alkyl group, namely, a trifluoromethyl group, as an example of A in his Examples and, it would be quite difficult, on the basis of this example alone, to obtain an idea that an alkoxy group would yield a polyimide of totally different properties.

6) Harris describes the possibility that A can be an alkoxy group. However, a large number of equally useful substituents other than the alkoxy groups are cited; such substituents include a vast number of groups such as substituted alkyl groups and substituted aryl groups and it is not easy to select alkoxy groups from them. Furthermore, since the number of carbon atoms in the alkoxy groups is not limited, this number itself would become quite large if isomers were included and it is not easy to select only those alkoxy groups which contain 3 to 6 carbon atoms from them. When a compound of interest happens to have an alkoxy group, one would normally first think of an alkoxy group containing one or two carbon atoms, but such alkoxy groups are not very

effective.

I, the undersigned petitioner, further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 of title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

July 29, 2009

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